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Quantum Dot Solar Cells with Multiple Exciton Generation

M.C. Hanna, M.C. Beard, J.C. Johnson, J. Murphy, R.J. Ellingson, and A.J. Nozik

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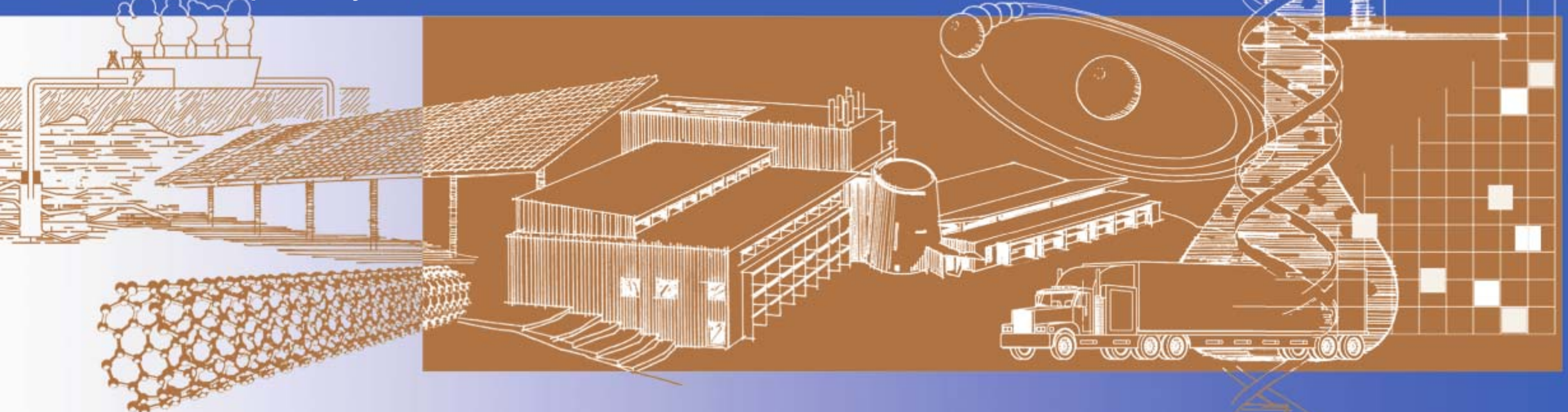

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ABSTRACT

We have measured the quantum yield of the multiple exciton generation (MEG) process in quantum dots (QDs) of the lead-salt semiconductor family (PbSe, PbTe, and PbS) using fs pump-probe transient absorption measurements. Very high quantum yields (up to 300%) for charge carrier generation from MEG have been measured in all of the Pb-VI QDs. We have calculated the potential maximum performance of various MEG QD solar cells in the detailed balance limit. We examined a two-cell tandem PV device with singlet fission (SF), QD, and normal dye (N) absorbers in the nine possible series-connected combinations to compare the tandem combinations and identify the combinations with the highest theoretical efficiency. We also calculated the maximum efficiency of an idealized single-gap MEG QD solar cell with M multiplications and its performance under solar concentration.

1. Objectives

The objective of this continuing project is to explore the possibility of greatly enhanced conversion efficiency in third-generation PV cells based on charge multiplication from the process of multiple exciton generation (MEG) in low-bandgap semiconductor quantum dots (QDs).

2. Technical Approach

We use fs pump-probe transient absorption dynamics to study carrier relaxation and MEG processes in QDs. For efficiency calculations, we use a detailed-balance model in which we assume complete light absorption for photon energy greater than the absorption threshold, charge multiplication occurs by MEG or singlet fission (SF), all photogenerated carriers are collected, and the only loss mechanism is due to spontaneous radiative recombination.

3. Results and Accomplishments

Extending our previous measurements of high MEG quantum yield in PbSe QDs, we found similar MEG quantum efficiencies in PbS and PbTe QDs. The quantum yield reached 300% at 4 x Eg, where Eg is the bandgap of the QD.

We have generalized our model for calculating the maximum efficiency of an idealized MEG QD solar cell to include M multiplications and the expected performance under solar concentration. For a given bandgap, Eg, the maximum number of excitons, Mmax, that can be produced per photon by MEG is dictated by energy conservation considerations. The value of Mmax is given by Mmax = Emax/Eg, where Emax ~ 4.4 eV is the highest photon energy in the AM1.5 solar spectrum. As an example, for Eg = 0.75 eV, Mmax = 5, as shown in Fig. 1.

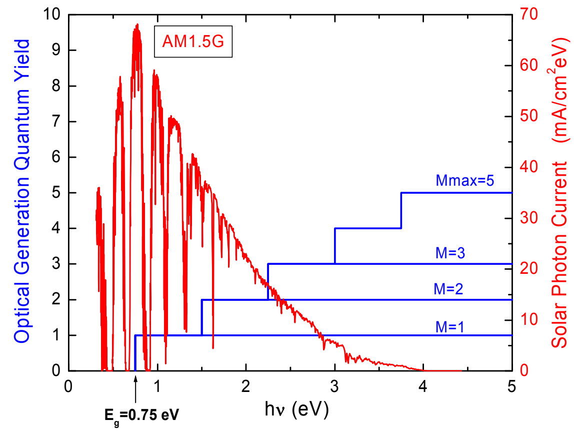

Fig. 1. MEG quantum yield models for Eg = 0.75 eV overlapping the AM1.5G spectrum. The maximum possible multiplication for this bandgap is Mmax = 5.

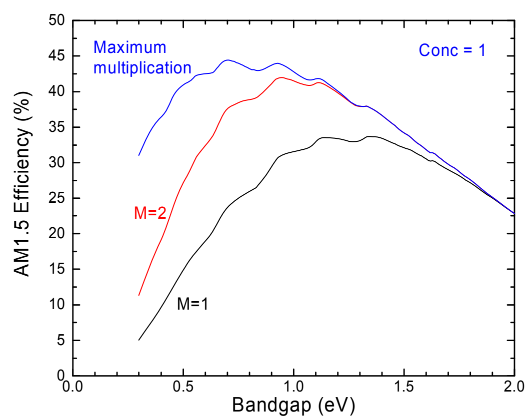

Fig. 2. Maximum efficiency at one sun for a MEG QD solar cell versus bandgap for multiplications of M = 1, 2, and Mmax.

The calculated efficiency versus bandgap for a MEG cell with M = 1, 2, and Mmax under one sun concentration is shown in Fig. 2. Allowing the maximum possible multiplication in a MEG cell raises the ultimate efficiency only a small amount compared to M = 2 (from 42.0% at Eg = 0.95 eV to 44.4% at Eg = 0.7 eV). In general, cells with lower bandgap benefit more from allowing maximum multiplication. Figure 3 shows how the maximum efficiency of a MEG cell with maximum multiplication changes with concentration. The maximum possible efficiency at maximum multiplication is 84.9% at a bandgap of 0.05 eV.

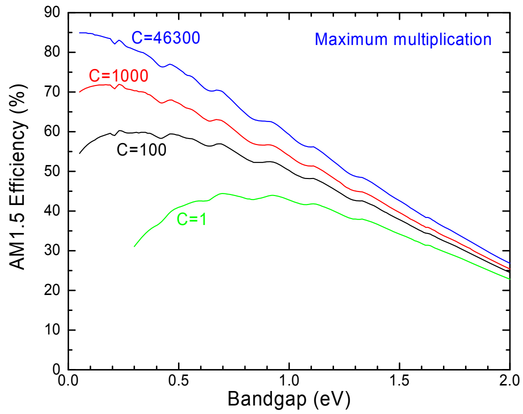

Fig. 3. Maximum efficiency versus bandgap of a MEG QD cell with maximum allowable exciton multiplication for different solar concentrations.

We have calculated the upper limits on the maximum efficiency of a two-gap tandem PV device with various combinations of a MEG QD absorber, a normal dye absorber (N), or an SF absorber as the top and bottom cells to identify the combinations with the highest theoretical efficiency. The efficiencies were calculated in the detailed-balance limit with the top and bottom cells electrically and optically in series.

The N-absorber generates one electron-hole (e-h) pair per photon absorbed above the bandgap. The QD-absorber generates two e-h pairs for photons with energy greater than twice the bandgap and one e-h pair for photons with energy between one and two times the bandgap. An ideal SF absorber has a triplet state (E_T = E_1) and a singlet state (E_S = 2 x E_1). The SF-absorber absorbs all photons with energy above the singlet state at twice the triplet state energy (E_T = bandgap) and generates two electrons per absorbed photon for photons in this energy range.

The maximum overall efficiency, along with the optimum bandgaps of the top and bottom cells for each of the nine possible combinations, is summarized in Table 1. The best absorber combinations, (SF,QD), (QD,N), (QD,QD), and (QD,SF), have a QD-absorber as the top cell or the bottom cell. These cells have a potential efficiency of greater than 47% for one sun illumination, which exceeds the value of 45.7% for the usual (N,N) tandem.

Table 1. Maximum AM1.5G theoretical efficiency for SF, QD, and N tandem cell combinations at the optimum top and bottom cell bandgaps.

Top/Bottom Absorber	Top Cell Bandgap (eV)	Bottom Cell Bandgap (eV)	Maximum Efficiency (%)
(N,N)	1.63	0.95	45.7
(N,QD)	1.61	0.95	45.7
(N,SF)	1.39	0.48	43.8
(QD,N)	1.63	0.95	47.1
(QD,QD)	1.46	0.68	47.6
(QD,SF)	1.40	0.48	47.3
(SF,N)	0.95	0.95	45.4
(SF,QD)	0.84	0.70	47.7
(SF,SF)	0.82	0.48	45.7

4. Conclusions

We have measured very high MEG quantum yields (up to 300%) for charge carrier generation in Pb-VI QDs (VI = S, Se, Te). We have calculated the maximum attainable efficiencies of both single-gap and two-gap tandem cells that incorporate MEG QD absorbers. Future work will focus on studying the MEG process in other QD materials, producing information on charge transfer and optical properties of QD/TiO_2 composites, and modeling the electrical behavior and potential performance of MEG QD solar cells.

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